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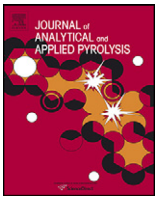

Reductive and oxidative combustion of polyethylene bags: Characterization of carbonaceous and nitrogenous species

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ABSTRACT

This study aims to experimentally characterize the gaseous carbonaceous and nitrogenous species from the reductive and oxidant combustion of polyethylene plastic bags. The experimental device used is the tubular furnace, coupled to two gas analyzers: a Fourier transform infrared analyzer (FTIR) and a non dispersive infrared analyzer (NDIR). The gaseous products analyzed are: CO, CO_2, CH_4, C_3H_8, C_2H_4, C_2H_2, C_6H_6, HCN, N_2O, NO, NO_2 and NH_3. The experiments were conducted at temperatures ranging from 800 to 1000 °C. The results obtained allow us to note that carbonaceous compounds are mainly emitted as carbon oxides (CO and CO_2) whether you are reductive combustion or oxidative combustion.

In addition:

- Under reductive conditions, combustion is controlled by oxygen. The hydrocarbon most active in the formation of carbon monoxide is ethylene (C_2H_4) and to a lesser extent, from 900 °C, acetylene (C_2H_2). The extents we have made show that ammonia seem to be emitted during combustion with 10% of oxygen.
- In an oxidative environment, there is production of C_6H_6 in substantial quantities, which partly explains the presence of soot and tar in the smoke exhaust ducts. The C_2H_4, CH_4 and C_2H_2 are hydrocarbons most active in the formation of CO and CO_2. Increasing of concentration of local oxygen from 10 to 21% for the combustion of plastic bags, favors an increase in efficiency of carbon conversion about 30%. About 99% of the carbon of the fuel is found to be converted to carbon oxides or hydrocarbons. Nitrogen monoxide (NO) is the major component among the gases measured with a conversion rate of nitrogen about 20%, three times larger than that obtained during the reductive combustion of plastic bags with 10% oxygen.

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1. Introduction

Nowadays, the urban waste management has become a major problem in most of the towns. Indeed, the rate of citizens is increasing all over the world, inducing an increase of the production of urban wastes. The composition of these wastes is very complex, depending on the town, the type of climate, the standing of population... Plastics bags are present in the waste and need to be selectively managed because of their environmental and health impact. Indeed, these used plastic bags cause damages for agriculture, breeding, and generate also visual pollution in the towns. These synthetic plastic bags are not biodegradable and represent a long term problem for environment managers. Recycling and landfill of plastic bags is not always easy and/or cost effective. Under controlled conditions, thermal treatments can be considered as efficient and reliable methods to reduce the size and the volume of wastes containing plastic bags. However, this incineration generates both solid and gaseous pollution which need to be reduced.

Many authors worked on the characterization of the gas from incineration units [1–19]. It appears that many toxic compounds such as carbon, sulfur, and nitrogen oxides, chlorine products (dioxins and furans), volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), dust... are present in gaseous emissions resulting from incineration [1–19]. Many other authors worked specially on the characterization of reactional intermediates during the combustion process [1–19]. Unfortunately most of

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these authors did not compare the reactivity of the reactional intermediates for the formation of carbon oxide. Only Faravelli et al. [14], reported the formation of CO by oxidation of ethylene.

The aim of our study is to characterize the gas after pyrolysis, reductive combustion and combustion, and to establish some mechanisms of formation of carbon monoxide from hydrocarbon reactional intermediates.

A study of characterization of gaseous carbonaceous and nitrogenous species produced during combustion of plastic bags made from polyethylene, under experimental conditions close to the incineration of plastic materials, was conducted. These experimental studies allow identification of the main reaction pathways of degradation of plastic bags and determining the composition of gas of burning. Our objective is to characterize, under reductive and oxidative environment, gases from the combustion of polyethylene plastic bags. To do this we have do the combustion, respectively with 10% and 21% of oxygen, using the tubular furnace. The term "reductive combustion" is explained by the fact that we reduce the rate of oxygen in the air used in combustion. The device of the tubular furnace is coupled to two analyzers of gas: an analyzer Fourier transform infrared (FTIR) and a non-dispersive infrared analyzer (NDIR). The fuel is mainly composed of carbon, oxygen, hydrogen and nitrogen. Some authors [1–7], have shown that the species analyzed (CO, CO_2, CH_4, C_3H_8, C_2H_4, C_2H_2, C_6H_6, HCN, N_2O, NO, NO_2 and NH_3·HCN and NH_3), are main reactional intermediates for the formation of NO of fuel, and others [2,5–10] confirm, through their results, that the NO_2 and N_2O are involved in the reduction of NO. Similarly, work on the precursors of formation of CO and CO_2, led by [11–14], show that hydrocarbons are the precursors to the formation of CO and CO_2 and intervene in the process of formation and destruction of NO. This work will identify, depending on the temperature, the volatile species emitted, changes in the rate of conversion of carbon and nitrogen in each species analyzed and to understand how these compounds contribute to the formation of major pollutants such as nitrogen and carbon oxides.

2. Experimental part

2.1. Properties of fuels

An elementary analysis of polyethylene bags was performed to accurately characterize our fuel and carry out material balances. To do this, samples of each component were analyzed for the determination of the majority elements i.e.: C, H, O, N, S, Cl, H_2O and ashes. These tests have been carried out by the analysis of the National Center for Scientific Research (CNRS) with 5 different samples of each fuel. The values presented in Table 1 correspond to the average of rate from 5 analyses.

The polyethylene bags are made from PE granules, but during the manufacturing process, some molecules can be added to modify the physical properties of the bags. Thus, we have made in Table 2, a comparison of elementary analysis of the polyethylene bags and the pure polyethylene [15–18].

Table 1
Elementary composition of plastic bags.

Elements	Polyethylene bags
Carbon (C)	73.8%
Hydrogen (H)	11.5%
Nitrogen (N)	0.2%
Sulphur (S)	0.2%
Oxygen (O)	4.8%
Chlorine (Cl)	300 ppm
Water (H_2O)	2.5%
Ashes	6.7%

Table 2
Comparison of elemental analysis of the pure polyethylene and plastic bags.

Elements	Polyethylene bags	Pure Polyethylene
Carbon (C)	73.8%	85.4–86.4%
Hydrogen (H)	11.5%	13.5–14.3%
Nitrogen (N)	0.2%	0%
Sulphur (S)	0.2%	0–0.08%
Oxygen (O)	4.8%	0–0.2%
Chlorine (Cl)	300 ppm	0 ppm
Water (H_2O)	2.5%	–
Ashes	6.7%	0%

In view of this review, we find that the polyethylene bags have a chemical composition different from that observed with the pure polyethylene. The differences are mainly at the level of carbon, oxygen, chlorine and ash. These differences are related to the addition of adjuvant.

2.2. Description of experimental device

The device used is the tubular reactor instrumented to study the thermal degradation of solid materials in pyrolysis and combustion. This device was used to determine for different temperatures, the composition of the volatile gases generated by fast pyrolysis. Fig. 1 presents a diagram of the tubular reactor used.

The central part of the diagram consists of the tubular kiln and the quartz reactor. The kiln is placed horizontally and measure 88 cm long. It is connected to an automatic temperature control (heat resistance) with 1250 °C like maximum instruction. The quartz reactor with a total length of 120 cm and an inside diameter of 7 cm, is partly placed inside the kiln. A thermocouple of type K is placed at its center in order to know the temperature. A basket sample holder (in quartz) is used for the introduction of the fuel inside the kiln.

The flow of oxidizer (nitrogen, "nitrogen + air" or air, depending on the chosen degradation type) is performed by a digital control

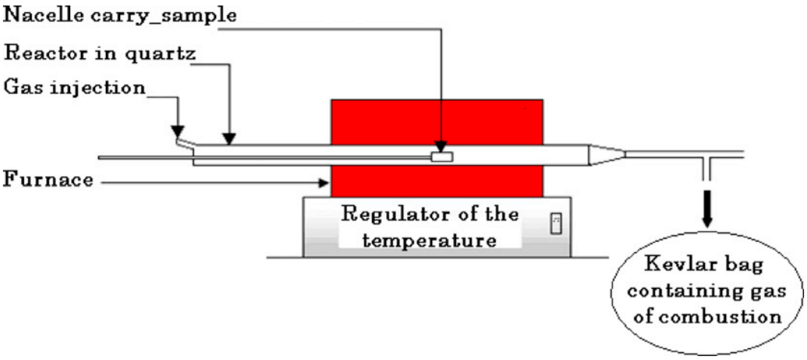
Nacelle carry_sample
Reactor in quartz
Gas injection
Furnace
Regulator of the temperature
Kevlar bag containing gas of combustion

Fig. 1. Device of tubular furnace.

Table 3
Conditions of thermal degradation tests.

Type of dégradation	Carrier gas	Sample mass (mg)	Temperatures (°C)	Flow of carrier gas (Nl/min)	Residence time (s)
Combustion (10%)	Air + Nitrogen	500	800	11.2	2
			850	10.7	
			900	10.2	
			950	9.8	
			1000	9.4	
Combustion (21%)	Air	500	800	11.2	2
			850	10.7	
			900	10.2	
			950	9.8	
			1000	9.4	

box (ELECTRONIC CONTROLLER 0154) linked to a mass flowmeter of brand BROOKS, type 5850E that allows a maximum flow gas of 30 Nl mn^{-1} with a precision of 10^{-1} Nl mn^{-1}. A fraction of gas from the degradation is directly collected in a Kevlar bag with a capacity of 37 L. The volume of gas collected is calculated by the flow of gas carried and the time of filling the bag. Before each filling, the bag is cleaned and the vacuum is made by aspiration with a pump. The gas collected is then taken continuously with a pump and sent to the gas analyzer with a flow rate of 2 L mn^{-1}. These gases pass through a filter that removes soot and tars. Then they cross a box, by Pelletier effect, which removes water vapor before analysis.

The gases collected in the bag are spread into series toward two types of analyzers

- Infrared non dispersive (IRND) and paramagnetic analyzers, of brand Servomex, to measure the concentrations of CO, CO_2, O_2, SO_2, and NO.
- FTIR spectrometer brand Thermo Optek NICOLET, type AVATAR, to determine the concentrations of CO, CO_2, CH_4, C_2H_2, C_2H_4, C_2H_6, C_3H_8, C_6H_6 and NH_3, HCN, NO, NO_2, N_2O and SO_2.

2.2.1. Infrared non-dispersive and paramagnetic analyzers

A suction pump allows the circulation of a flow of smoke of 2 Nl mn^{-1}. The accuracy of the analyzers is of 2% of reading for the analysis of O_2, SO_2 and CO_2, and 5% of the value read for the analysis of CO and NO [20]. The detection limit for NO, CO and SO_2 is 10 ppm and 0.1% for O_2 and CO_2 [21]. The response time of the analyzers is 30 s.

2.2.2. Fourier transform infrared analyzer (FTIR)

Infrared spectrometer used has been established at the Mine school of Albi (France) by [21] in its work for doctoral degrees. Since [22–28], have used this apparatus with this quantification process, for their research works. For qualitative analysis we can say that because of the weak interaction between the molecular gases, the spectrum of a mixture of gases can be considered as a set of pure compound spectra. This allows for two types of product identification in the spectra analysis:

- by visual identification method: this method is to use spectra of pure compounds contained in a commercial library (Aldrich, Sigma...);
- method of personal identification: identification is made by visual layering of strips characteristic of each compound in the spectrum of reference recorded by analysis of standard gases.

The flow of gas through the cell is 2 Nl mn^{-1}. Measurement accuracy is about 5% of reading for most gases quantified, except for CO_2, for which the accuracy is reliable only for a concentration of less than 1% i.e. 10^4 ppm. After, quantification is very uncertain and we take the value indicated by the analyzer non-dispersive infrared. The threshold for detection of NO, N_2O and NH_3 is 10 ppm [21]. The response time of the FTIR is 40 s.

2.3. Experimental conditions

The polyethylene bags are cut into square sheets of 1.5 cm of side with less of 1 mm thick. Samples, with a mass of 500 mg, were then weighed with a precision balance, 10^{-2} mg. These sizes and masses allow avoiding the effect of temperature gradient at the degradation of the fuel. This physical phenomenon influences the mode of degradation. Indeed, the degradation of a "fuel thermally thick" would imply the presence of a temperature gradient, while that of a "fuel thermally thin" would be no temperature gradient since in this case the temperature is supposed uniform throughout the solid.

The temperature of preheating of the kiln is programmed to the desired value. The flow of gas is selected to have a fixed residence times. It is based on the residence time of hot gas and the local temperature of degradation. Once the set temperature is reached and stable, the oxidizing gas flow is set. The flow of oxidizing gas is performed by a digital control box, brand ELECTRONIC CONTROLLER, type 0154, connected to a mass flowmeter brand BROOKS, type 5850E, which allows a maximum flow of gas 30 Nl mn^{-1} with accuracy to 10^{-1} Nl mn^{-1}.

Then, the sample is introduced in the kiln with a basket quartz sample holder. The basket is manually entered into the kiln at a constant speed. It is at the center of the kiln, where the temperature is given by a thermocouple of type K. The gases from the decomposition are transported to the gas analyzer. Before each new test the basket is beforehand removed from the kiln. The response time of the analyzer FTIR and the analyzers SERVOMEX are high (respectively 40 and 30 s), the gases from the thermal degradation are stored in a bag of Kevlar of 37 L of capacity. In other words, flow and pressure of gas leaving the reactor are very high for the analyzers which save different concentrations of the gaseous species. The gases collected are then continuously take with a pump (with a rate of 2 Nl mn^{-1}) and sent to the gas analyzers. These gases pass through a filter that removes soot and tars in order to prevent their deposit on cells and/or on other internal components of the analyzers. These deposits can damage the components. The transfer lines of gases, the reactor to the bag and the bag to the analyzers are cleaned (by blowing) after each test, to remove residues of soot and tar. The bag for the collection of the thermal degradation gases is changed every five trials, this will prevent deposits of tar, soot and/or particles that can compromise the reliability of the results by reacting with the gases. The results are treated by computer in real time. Table 3 shows the various conditions for which the tests were doing.

Our temperatures conditions correspond to those used in the thermal degradation of cellulosic and plastic materials [19,29–34]. The residence time of gases in the hot zone of the reactor was chosen so that the airflow is not too important: given the mass of each

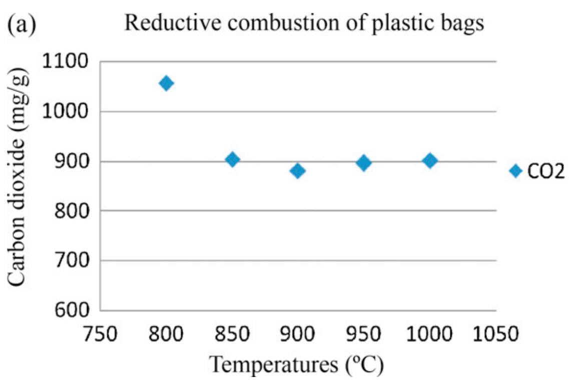

Fig. 2. (a) Rate of carbon dioxide analyzed in the reductive combustion of plastic bags as a function of temperature. (b) Rate of carbon monoxide analyzed in the reductive combustion of plastic bags as a function of temperature.

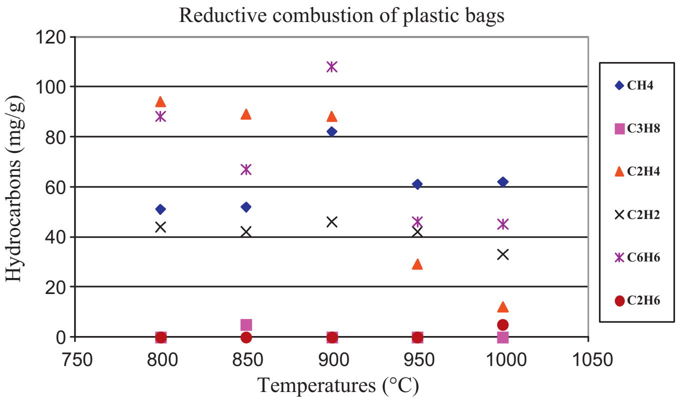

Fig. 3. Rate of hydrocarbons analyzed during combustion reductive of plastic bags as a function of temperature.

sample, a flow too high would imply an only partial degradation of fuel. Indeed, an air flow promotes a very significant cooling of the reaction fuel and a residence time of hot gas too short, and this would cause a decrease in temperature of degradation and an incomplete degradation of fuel.

3. Results and discussion

3.1. Reductive combustion

3.1.1. Carbon

In Figs. 2a and 3, emissions from carbonaceous species analyzed, depending on the temperature during reductive combustion (10% of oxygen) in polyethylene plastic bags, are presented. Table 4 gives the conversion rates of volatile carbon in all compounds analyzed.

Table 4
weight percentage of carbon conversion during reductive combustion of plastics bags as a function of temperature.

Percentage of volatile carbon, in each compound					
	800	850	900	950	1000
CO	8%	7%	8%	8%	9%
CO_2	39%	33%	33%	33%	33%
CH_4	5%	5%	8%	6%	6%
C_3H_8	0%	1%	0%	0%	0%
C_2H_4	11%	11%	10%	4%	2%
C_2H_2	6%	5%	6%	5%	4%
C_6H_6	11%	8%	14%	6%	6%
C_2H_6	0%	0%	0%	0%	1%
Total	80%	70%	79%	62%	61%

The compounds mainly emitted are carbon dioxide (CO_2) and carbon monoxide (CO), whatever the temperature of combustion. In Fig. 2a and b, when the temperature increases, there is initially a decrease and then stabilization of CO_2 emissions; while the CO increases linearly from 850 °C. This phenomenon is due to the decrease of concentration of local oxygen when the temperature increases. Then, the increase of CO can be justified by the assumption that the oxidation reactions of hydrocarbons into CO are faster than those of CO into CO_2 [35,36]. Looking at Fig. 3 we see that ethylene (C_2H_4), methane (CH_4), benzene (C_6H_6) and acetylene (C_2H_2) are produced in smaller quantities but not negligible. This result is due to insufficient oxygen to generate the complete oxidation of hydrocarbons. Propane (C_3H_8) and ethane (C_2H_6) are virtually nonexistent. Comparing with the results of pyrolysis [29], it seems that the most active hydrocarbon in the formation of carbon monoxide is ethylene (C_2H_4) and acetylene (C_2H_2) from 900 °C.

3.1.2. Nitrogen

The balance (in mg g^{-1}) of nitrogenous species and the rate of conversion of nitrogen in each gaseous compound, for combustion with 10% of oxygen, plastic bags between 800 and 1000 °C with a residence time of 2 s, are presented respectively in Fig. 4 and Table 5.

- The measures we have made show that ammonia seems to be emitted during combustion in a reductive environment, with 10% of oxygen. These levels represent an efficiency of nitrogen conversion from 3 to 7%. It may be that:
  - The presence of oxygen allows the reduction of other compounds into the NO and NO into N_2. However, an analysis of N_2 could not be performed.

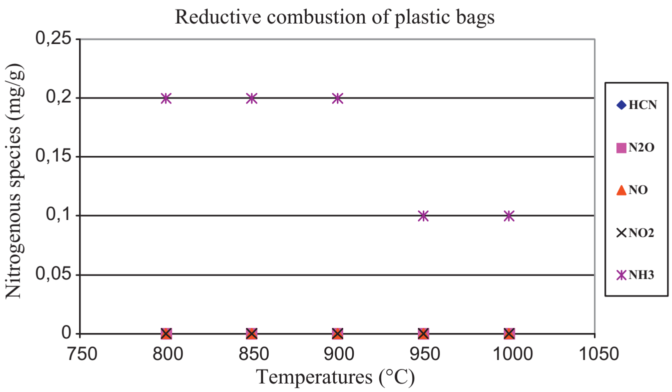

Fig. 4. Rate of nitrogenous species analyzed in the reductive combustion of plastic bags as a function of temperature.

Table 5
Mass rate of nitrogen detected during the reductive combustion of plastic bags.

Percentage of volatile nitrogen, in each compound					
	800	850	900	950	1000
HCN	0%	0%	0%	0%	0%
N_2O	0%	0%	0%	0%	0%
NO	0%	0%	0%	0%	0%
NO_2	0%	0%	0%	0%	0%
NH_3	7%	7%	7%	3%	3%
Total	7%	7%	7%	3%	3%

- Other nitrogenous compounds have been emitted and the values emitted are below the detection limit of the analyzers.

Table 6
Weight percentage of carbon conversion during oxidant combustion of plastic bags as a function of temperature.

Percentage of volatile carbon, in each compound					
	800	850	900	950	1000
CO	9%	10%	11%	14%	12%
CO_2	69%	69%	70%	71%	78%
CH_4	2%	2%	3%	4%	2%
C_3H_8	0%	0%	0%	0%	0%
C_2H_4	4%	3%	2%	1%	0.4%
C_2H_2	3%	3%	3%	3%	2%
C_6H_6	11%	11%	10%	7%	4%
C_2H_6	0%	0%	0%	0%	0.8%
Total	98%	98%	99%	100%	99.2%

3.2. Oxidant combustion

3.2.1. Carbon

The curves of Figs. 5 and 6 correspond to emission rates of carbon oxides and hydrocarbons, depending on the temperature during the oxidative combustion of plastic bags made from polyethylene. Table 6, gives a balance of the percentage of conversion of carbon in each gas analyzed.

The oxidative combustion of plastic bags, with 21% of oxygen, revealed that for all temperatures studied, the CO_2 is emitted predominantly (average 1900 mg g^{-1}), followed by CO (average 198 mg g^{-1}). Also included in the measured gases, CH_4, C_2H_4, C_2H_2 and C_6H_6. The presence of C_6H_6 may explain the presence of soot and tar in the smoke exhaust ducts. This result means that the degradation of PE releases heavy hydrocarbons whose oxidation is slow. We do not detect C_2H_6 and C_3H_8 in the smoke analyzed in our experimental conditions. Compared with levels of gases measured during combustion at 10% of oxygen, hydrocarbon emissions are decreasing, while the level of CO and CO_2 increases. It seems that the C_2H_4, CH_4 and C_2H_2 are hydrocarbons most active in the formation of CO and CO_2, because being reduced by over 50% compared to emission rates recorded during reductive combustion with 10% of oxygen, this was confirmed by [37]. On average, 70% of the volatile carbon is converted to CO_2, 11% into CO and 8% into C_6H_6. For the other three hydrocarbons (C_2H_4, CH_4, C_2H_2), the conversion rate of volatile carbon is less than 4%. The increase in the rate of oxygen to 21%, for combustion of plastic bags, favors an increase in the rate of carbon conversion to CO_2 by over 50% and an increase in the total efficiency of combustion nearly 30%. Thus, approximately 99% of the carbon of the fuel is found converted into oxides of carbon or into hydrocarbons.

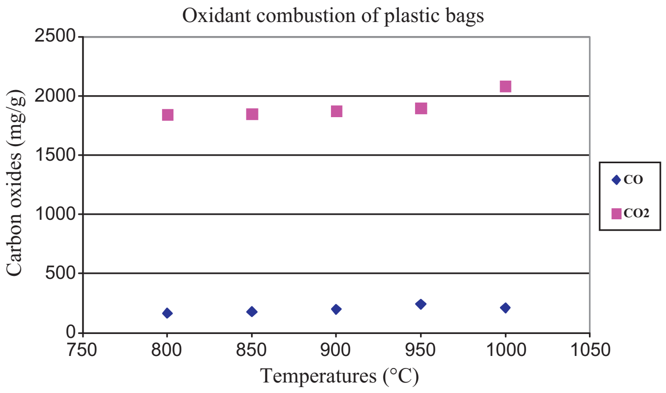

Fig. 5. Rate of carbon oxides analyzed in the oxidant combustion of plastic bags as a function of temperature.

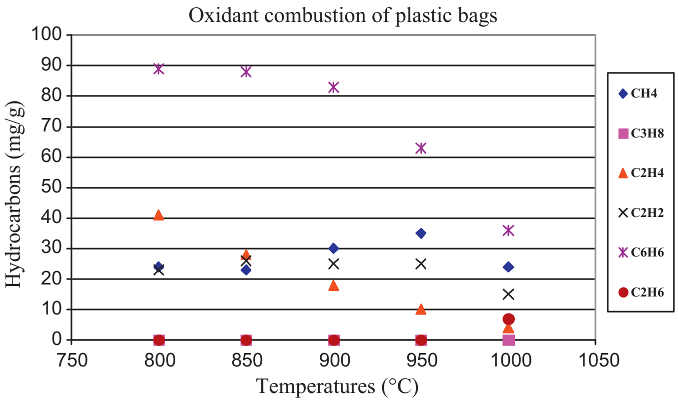

Fig. 6. Rate of hydrocarbons analyzed during combustion oxidant of plastic bags as a function of temperature.

3.2.2. Nitrogen

Fig. 7 gives the concentrations of nitrogenous species analyzed in the oxidative combustion of plastic bags, for temperatures ranging from 800 to 1000 °C and a residence time of 2 s. Under the same conditions, Table 7 presents the mass ratio (%) of nitrogen conversion in all compounds analyzed. The monoxide of nitrogen (NO) is the major component among the measured gases. Unlike the case of combustion at 10% of oxygen, there is practically no NH_3. The presence of HCN to the detriment of NH_3 can be justified by the fact that the formation of NO from the compounds of types NHi• and HNO• is preferred [8–10]. In terms of efficiency of conversion of fuel nitrogen, the total rate of conversion rate is around 20%. Thus, for an oxidative combustion with 21% of oxygen, we have an efficiency of conversion that is three times that

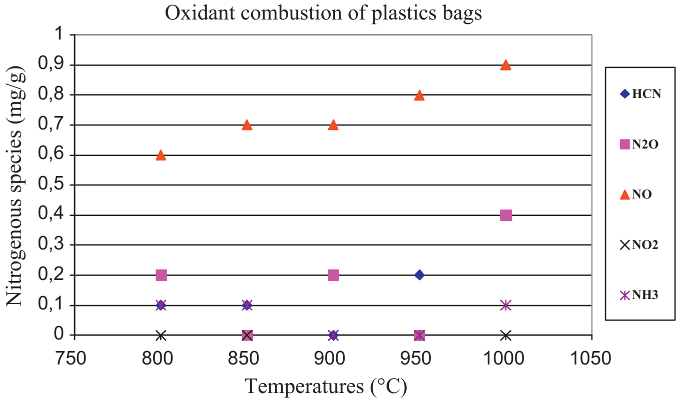

Fig. 7. Rate of nitrogenous species analyzed in the oxidant combustion of plastic bags as a function of temperature.

Table 7
Mass rate of nitrogen detected during the oxidant combustion of plastic bags.

	Percentage of volatile nitrogen, in each compound				
	800	850	900	950	1000
HCN	2%	3%	0%	6%	11%
N_2O	5%	0%	5%	0%	13%
NO	15%	17%	16%	19%	22%
NO_2	0%	0%	0%	0%	0%
NH_3	2%	3%	0%	0%	2%
Total	24%	23%	21%	25%	48%

obtained in the reductive combustion of plastic bags, with 10% of oxygen.

4. Conclusion

Our study aims to characterize carbonaceous and nitrogenous species during combustion oxidative and reductive of plastic bags. This characterization is needed to better understand the mechanisms of formation of gaseous pollutants. It was carried out using the device the tubular furnace at temperatures ranging from 800 to 1000 °C. Reductive combustion tests (10% of oxygen) and oxidative combustion tests (21% oxygen) were made. The kiln is connected to two gas analyzers: spectrometer FTIR and a NDIR. The gases analyzed were: CO, CO_2, CH_4, C_3H_8, C_2H_4, C_2H_2, C_6H_6, HCN, N_2O, NO, NO_2 and NH_3. The results obtained in this study can make the following balance:

- In reductive environment, combustion is controlled by oxygen. The carbonaceous compounds are mainly emitted as carbon oxides (CO and CO_2). The hydrocarbon most active in the formation of carbon monoxide is ethylene (C_2H_4), and acetylene (C_2H_2) to a lesser extent from 900 °C. The measures we have made show that ammonia only, seem to be emitted during combustion in a reducing environment, with 10% of oxygen.
- In oxidative combustion with 21% of oxygen, CO_2 and CO are the compounds mainly emitted. The C_6H_6 is products in substantial quantities, which partly explains the presence of soot and tar in the smoke exhaust ducts. The C_2H_4, CH_4 and C_2H_2 are hydrocarbons most active in the formation of CO and CO_2. Increasing the local concentration of oxygen from 10 to 21% for the combustion of plastic bags, favors an increase of total efficiency of carbon conversion by almost 30%. Thus, approximately 99% of the carbon of the fuel is found be converted into oxides of carbon or hydrocarbons. The monoxide of nitrogen (NO) is the major component among the gases measured, with a conversion rate of nitrogen about 20%, three times larger than that obtained during the reductive combustion of plastic bags with 10% of oxygen.

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